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Jul 8, 1997

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TITLE: Copper@ impregnated graphite material - for use in plasma nuclear fusion reactor, and spacecraft and aircraft members

PATENT-ASSIGNEE: TOYO TANSO CO (TOTAN)

PRIORITY-DATA: 1995JP-0315738 (October 27, 1995)

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## APPLICATION-DATA:

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INT-CL (IPC): C04 B 35/52; C04 B 41/88; G21 B 1/00

ABSTRACTED-PUB-NO: JP09175877A

## BASIC-ABSTRACT:

Copper impregnated graphite material comprises at least 70 volume % open porosity texturally dense and isotropic graphite material, having a bulk density of at least 1.76 Mg/m-3, with 5 - 18 volume % having a mean pore radius of 0.1 - 2.5  $\mu$  m, measured by a mercury method. The material is impregnated with copper alloy, containing 1 - 7 weight % of at least one element from the group with a graphite-copper reaction standard generation enthalpy up to -50 kJ/mole and the remainder copper. The final material has a bulk density of at least 3 Mg/m-3 after impregnation.

USE - The material is used for coating areas contacting plasma in a plasma nuclear fusion reactor, and for coating space and aircraft members.

ADVANTAGE - The graphite material is uniformly and isotropically impregnated with copper.

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EQUIVALENT-ABSTRACTS:

CHOSEN-DRAWING: Dwg.0/1

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**KURODA KOJI**  
**SOGABE TOSHIKI**

(54) **COPPER-IMPREGNATED GRAPHITE MATERIAL**

## (57) Abstract:

**PROBLEM TO BE SOLVED:** To obtain a lightweight copper-impregnated graphite material having a homogeneous macrotexture, increased denseness, and improved handleability by impregnating an isotropic graphite material with a specific copper alloy on its open pores.

**SOLUTION:** An isotropic graphite of high dense texture with a bulk density of  $21.76 \text{ Mg/m}^3$ , an average pore diameter of 5-18vol.% and an average pore radius of

0.1-2.5 $\mu\text{m}$ , measured according to the mercury porosimetry and an alloy of copper with 1-7wt.% of at least one selected from the elements having 2-50kJ/mole of standard enthalpy of formation according to the reaction with graphite and copper are charged in the vessel, heated over the melting point of the copper alloy, pressurized under several Mpa-150Mpa for 1-60 minutes to impregnate the pores in the graphite material with the copper alloy over 70vol% of the pore volume whereby the objective copper-impregnated graphite material of  $23.0 \text{ Mg/m}^2$  bulk density is obtained.

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(33) 優先権主張国	日本 (J P)		

(54) 【発明の名称】 銅含浸黒鉛材

(57) 【要約】

【目的】 本発明は、黒鉛材料中に銅をむらなく含浸でき、含浸作業時の取扱性にも優れた銅含浸黒鉛材料を提供することを目的とする。

【構成】 本発明に係る銅含浸黒鉛材は組織的に緻密な黒鉛材の開気孔に、銅と黒鉛及び銅との標準生成エンタルピーが何れも低い金属との合金を含浸することにより、取扱性に優れた銅がむらなく含浸されている。

## 【特許請求の範囲】

【請求項1】 かさ密度が $1.76 \text{ Mg/m}^3$  以上であり、そして水銀圧入法により測定した開気孔率及び平均気孔半径が夫々5乃至18体積%及び0.1乃至 $2.5 \mu\text{m}$ である組織的に緻密な等方性黒鉛材の開気孔の70体積%以上を、黒鉛及び銅との反応による標準生成エンタルピーが夫々1モルあたり $-50 \text{ kJ}$ 以下である元素群から選ばれる少なくとも1種の元素1乃至7重量%及び残部が実質的に銅から成る銅合金により含浸処理され、しかも含浸後のかさ密度を $3.0 \text{ Mg/m}^3$  以下としたことを特徴とする銅含浸黒鉛材。

## 【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明はプラズマ核融合炉のプラズマ対向部材や宇宙航空機用部材など高温に曝される構造部材として適した銅含浸黒鉛材に関する。

【0002】

【従来の技術】 黒鉛は金属に比べて軽量（真密度約 $2.16 \text{ Mg/m}^3$ ）で、 $3000^\circ\text{C}$ を超える温度でも融解、分解を起こさず、しかも高温になる程寧ろ機械的強度が高まるなど、高温材料として優れた特性を発揮するが、この黒鉛の性質を更に改良するために銅あるいはその他の金属を黒鉛に含浸することが行われている。とりわけ銅の黒鉛材料への含浸は、電気伝導性、熱伝導性の向上、しゅう動性の向上、機械的特性の改善、ガス透過の低減などを目的として、広く工業的に実施されている。

【0003】 しかしながら、一般的に銅融液は黒鉛に対してぬれが悪く、気孔中に含浸した際に気孔壁に十分密着せず、微視的に含浸むらが生じ、その結果上述の特性改良が十分に達成できないという問題点があった。

【0004】 含浸むらをなくす1つの方策として、特開昭51-24525号公報には、焼成炭素材への含浸に、ぬれ剤としてTi、Zr又はSiを添加した銅合金を用いることが教示されている。

【0005】 また、これよりも関連性は薄い、特開昭61-136644号公報には、含浸金属のぬれ性改良剤としてある種の金属の塩を用い、あらかじめこの金属塩を含浸し熱分解して金属を気孔壁に付着させた後に、例えばアンチモンやアンチモン銅を含浸する金属含浸炭素すり板材の製造が教示されている。

【0006】

【発明が解決しようとする課題】 しかしながら、上記特開昭51-24525号公報に具体的に記載されている銅含浸炭素材は、 $1200 \sim 1500^\circ\text{C}$ 程度で焼成されたかさ比重の低い所謂焼成ブロック材にしゅう動性を与えるために比較的多量の銅（融点約 $1083^\circ\text{C}$ ）を含浸したものであり、炭素材料の軽量高温材料としての特長が損なわれてしまう。しかも、含浸温度を低下させるためと、しゅう動性を高めるためにSn、Sb等を添加す

るが、これらはしゅう動性以外の特性の改良には寧ろマイナスとなる可能性もある。また、Tiは炭素との親和性が相対的に高過ぎるため、含浸後の含浸金属材と炭素材との分離を困難にさせるという不都合があった。

【0007】 本発明者らは、上記従来技術の問題点を解決し、緻密、軽量で、しかも取扱性に優れた銅含浸黒鉛材を得るべく鋭意検討の結果、均質なマクロ組織を有し緻密で高温材料としてとりわけ優れた特性を発揮する等方性黒鉛を用いた場合も、銅及び黒鉛との親和性が何れも高いある種の元素群を銅の添加剤として選択使用することにより、銅をむらなく含浸でき、軽量高温材料としての特長を保ちながら、取扱性にも優れた銅含浸黒鉛材が得られることを見出し、本発明を完成するに至った。

【0008】

【課題を解決するための手段】 即ち、本発明は、かさ密度が $1.76 \text{ Mg/m}^3$  以上であり、そして水銀圧入法により測定した開気孔率及び平均気孔半径が夫々5乃至18体積%及び0.1乃至 $2.5 \mu\text{m}$ である組織的に緻密な等方性黒鉛材の開気孔の70体積%以上を、黒鉛及び銅との反応による標準生成エンタルピー（ $\Delta H^\circ$ ）が夫々1モルあたり $-50 \text{ kJ}$ 以下である元素群から選ばれる少なくとも1種の元素1乃至7重量%及び残部が実質的に銅から成る銅合金により含浸処理され、しかも含浸後のかさ密度を $3.0 \text{ Mg/m}^3$  以下としたことを特徴とする銅含浸黒鉛材である。

【0009】

【発明の作用】 本発明者らは黒鉛気孔中への銅の含浸のメカニズムを究明するため、最初に理論的な考察により仮説を立て、次いでこの仮説を実験により検証するという科学的な手法により検討を行なった。即ち、先ず、ある物質への異種物質の含浸の完全性が、含浸材、被含浸材相互の反応性、親和性と深く関係するという着眼に基づいて、銅及び黒鉛と各種元素との反応性、親和性を確認した。図1にこれら元素間の反応の標準生成エンタルピーを示した。図中のデータの出典はNorth-Holland社刊、Cohesion in Metalsに記載された数値データである。

【0010】 次いで、図中の各元素の中から、銅含浸の完全性を高められる助剤の候補が、黒鉛及び銅との反応の標準生成エンタルピーが何れも低い、即ち負の数値で、かつ絶対値も高い元素であるとの仮説を立て、実験による検証を試みた。

【0011】 その結果、図1のエンタルピー値と含浸のメカニズムとが以下に説明する様に関連を持つことが確認できた。

【0012】 即ち、例えばTiは黒鉛との反応性は高いが銅との反応性が低く、このためTi炭化物の生成やCuTiと黒鉛との界面接着力の面では有利であるが、反面表面層での炭化物の生成が優勢になるため、CuTi

の組織内奥への含浸が阻害されたり、含浸後の含浸金属材と黒鉛材との分離が困難になるなどの不都合が生ずる。これに対して、黒鉛及び銅との標準生成エンタルピーが何れも低いと、炭化物生成、界面接着力、含浸材の組織内奥までの浸透が良く、しかも含浸後の含浸金属材と黒鉛材との分離も容易となる。そして、これらの作用が得られる黒鉛及び銅との反応のエンタルピーのしきい値が、約 $-50\text{ kJ/mol}$ であることが分った。この条件に合う元素は、例えばSc、Y、Zr、La、Hfである。

【0013】この様な条件の含浸助剤を選択使用することにより、本発明で使用するかさ密度が $1.76\text{ Mg/m}^3$ 以上であり、そして水銀圧入法により測定した開気孔率及び平均気孔半径が夫々5乃至18体積%及び0.1乃至2.5 $\mu\text{m}$ である様な組織的に極めて緻密で、従って含浸が非常に困難な等方性黒鉛材へも銅を多量且つ組織内奥まで含浸でき、含浸率を70体積%以上としてもかさ密度が $3\text{ Mg/m}^3$ 以下という、軽量且つ緻密な組織の銅含浸黒鉛材が得られ、しかも含浸後の取扱性も良くなる。なお、含浸助剤の量を1~7重量%と規定したのは、1重量%未満では上記の様な作用が十分に得られず、7重量%を超すと含浸後の含浸銅合金と銅含浸黒鉛材との分離が困難になるためである。

【0014】

【発明の構成の詳細な説明】本発明で使用する等方性黒鉛材は、通常はコークス等の骨材にピッチ等のバインダーを加えて混練した後、冷間等方圧加圧成形を施し、焼成、黒鉛化に、必要に応じてピッチ含浸、再焼成、樹脂含浸、高純度化等の工程を経た、炭素のみから実質的に成る材料乃至は炭素を主成分とする材料から成り、ピッチ含浸品、樹脂含浸品等の含浸品を包含する所謂黒鉛化品等の等方性黒鉛材料を包含する。これらの等方性黒鉛材としては、特に異方比が1.2以下の等方性の高い黒鉛材料を用いることが好ましい。ここで、異方比が1.2以下であるとは、黒鉛材料における任意に直角をなす方向に図った固有電気抵抗の比の平均値が1.2以下であることを意味する。

【0015】本発明でいう等方性黒鉛材の平均気孔半径

は、例えば水銀圧入法により測定される累積気孔容積( $\text{m}^3/\text{Mg}$ )の $1/2$ に相当する半径値( $\mu\text{m}$ )として決定することができ、開気孔率は(かさ密度) $\times$ (全気孔容積) $\times 100$ で計算することができる。ここで、全気孔容積( $\text{m}^3/\text{Mg}$ )は圧力が予め定めた最高圧力、例えば98MPaまで達したときの累積気孔容積をいう。また、含浸率が70体積%以上であるとは、式： $I=100G/PD$ のI(%)で示される数値が70以上であることを意味する。但し、Pは等方性黒鉛材の開気孔の体積の実測値( $\text{m}^3$ )、Dは銅合金含浸材の真密度( $\text{Mg/m}^3$ )、Gは実際に含浸した銅合金の重量(Mg)を表す。つまり、I値は開気孔に占める含浸銅合金の体積割合を示す。

【0016】含浸は、例えば耐圧容器を用い、銅合金を炭素製のセラミック容器(るつぼ)に入れ、また等方性黒鉛材を炭素製のセラミック容器(サガー)に入れ、これらるつぼ、サガーを耐圧容器に装入し、次いで容器内を含浸銅合金の融点より高い温度に上げて加圧含浸する。加圧力は数MPa乃至150MPa程度、含浸時間は1~60分程度、好ましくは30乃至60分程度でよい。

【0017】なお、本発明において使用する銅合金の組成について、「残部が実質的に銅から成る」とは、残部を構成する銅に不可避免的な不純物が含まれていてもよいことを意味する。

【0018】

【実施例】

実施例1

東洋炭素(株)製の緻密質等方性黒鉛材(かさ密度 $1.82\text{ Mg/m}^3$ 、開気孔率13.7%、平均気孔半径 $1.5\mu\text{m}$ )を耐圧容器に収容し、 $1150^\circ\text{C}$ で溶融した7重量%のZrを添加した銅を $\text{N}_2$ ガスにて12MPaの圧力で1時間加圧含浸して銅含浸黒鉛材を得た。

【0019】含浸率、得られた銅合金含浸材のかさ密度、銅合金含浸材のガス透過率、及び含浸後の取扱性を表1にまとめた。

【0020】

【表1】

実施例 比較例 番号	含浸助剤	含浸率 (%)	かさ密度 ( $\text{Mg}/\text{m}^3$ )	ガス透過率 ( $\times 10^{-4}$ $\text{cm}^3/\text{s}$ )	含浸後の取扱性 (含浸材と被含 浸材との接着)
実施例 1	Zr 7%	89	2.84	4	接着は多少ある が取り出しは容 易である
対照例 1	なし	67	2.75	200	接着は全くない
比較例 1	Ti 1%	89	2.84	2	接着力が非常に 強く取り出しが 困難である

## 【0021】対照例1

銅にZrを添加しない以外は実施例1と同様にして銅含浸黒鉛材を得た。

## 【0022】比較例1

銅に1重量%のTiを添加した以外は実施例1と同様にして銅含浸黒鉛材を得た。

## 【0023】また、実施例1に係る本発明品、対照例

1、比較例1及び実施例1で使用了等方性黒鉛基材の室温、200℃、400℃及び800℃における熱伝導率をレーザーフラッシュ法で測定した。その結果を表2に示す。

## 【0024】

## 【表2】

処理 温度 (℃)	実施例 1	比較例 1	対照例 1	実施例 1 で 使用した 黒鉛基材
	銅-Zr合金	銅-Ti合金	銅	
室温	190	150	120	140
200	150	130	130	120
400	130	120	110	100
800	110	110	100	70

- 1) 熱伝導率はレーザーフラッシュ法で測定した。  
2) 単位は、 $\text{W}/(\text{m} \cdot \text{K})$ である。

【0025】CuをZrと合金化することによって、黒鉛材との接着性を向上させるだけでなく高い熱伝導率の黒鉛材を得られる。

## 【0026】

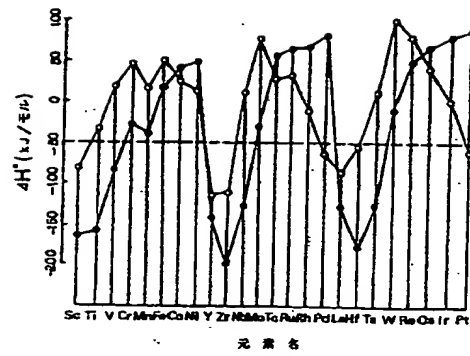
【発明の効果】本発明によれば、均質なマクロ組織を有し緻密で高温材料としてとりわけ優れた特性を発揮する等方性黒鉛を用いても銅をむらなく含浸でき、軽量高温

材料としての特長を保ちながら、緻密で、取扱性にも優れた銅含浸黒鉛材が得られる。

## 【図面の簡単な説明】

【図1】図1は各種元素と黒鉛及び銅との反応による標準生成エンタルピーを示した折れ線図である。図中●(黒丸)は黒鉛との反応によるものを示し、○(白丸)は銅との反応によるものを示す。

【図1】





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TITLE: Copper@ impregnated graphite material - for use in plasma nuclear fusion reactor, and spacecraft and aircraft members

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## PATENT-FAMILY:

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ABSTRACTED-PUB-NO: JP09175877A  
BASIC-ABSTRACT:

Copper impregnated graphite material comprises at least 70 volume % open porosity texturally dense and isotropic graphite material, having a bulk density of at least 1.76 Mg/m<sup>3</sup>, with 5 - 18 volume % having a mean pore radius of 0.1 - 2.5  $\mu$ m, measured by a mercury method. The material is impregnated with copper alloy, containing 1 - 7 weight % of at least one element from the group with a graphite-copper reaction standard generation enthalpy up to -50 kJ/mole and the remainder copper. The final material has a bulk density of at least 3 Mg/m<sup>3</sup> after impregnation.

USE - The material is used for coating areas contacting plasma in a plasma nuclear fusion reactor, and for coating space and aircraft members.

ADVANTAGE - The graphite material is uniformly and isotropically impregnated with copper.

ABSTRACTED-PUB-NO: JP09175877A  
EQUIVALENT-ABSTRACTS:

CHOSEN-DRAWING: Dwg.0/1

DERWENT-CLASS: E36 K05 L02 M22 X14  
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## (54) COPPER-IMPREGNATED GRAPHITE MATERIAL

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a lightweight copper-impregnated graphite material having a homogeneous macrotexture, increased denseness, and improved handleability by impregnating an isotropic graphite material with a specific copper alloy on its open pores.

SOLUTION: An isotropic graphite of high dense texture with a bulk density of  $\geq 1.76\text{Mg/m}^3$ , an average pore diameter of 5-18vol.% and an average pore radius of 0.1-2.5 $\mu\text{m}$ , measured according to the mercury porosimetry and an alloy of copper with 1-7wt.% of at least one selected from the elements having  $\leq 50\text{kJ/mole}$  of standard enthalpy of formation according to the reaction with graphite and copper are charged in the vessel, heated over the melting point of the copper alloy, pressurized under several Mpa-150Mpa for 1-60 minutes to impregnate the pores in the graphite material with the copper alloy over 70vol% of the pore volume whereby the objective copper-impregnated graphite material of  $\leq 3.0\text{Mg/m}^2$  bulk density is obtained.

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## LEGAL STATUS

[Date of request for examination]

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the copper sinking-in graphite material which was suitable as a structural member \*(ed) by elevated temperatures, such as a plasma opposite member of a plasma nuclear fusion reactor, and a member for aeronautics-and-astronautics machines.

[0002]

[Description of the Prior Art] Although that a graphite is lightweight (true-density about 2.16 Mg/m<sup>3</sup>) compared with a metal, and a mechanical strength increases rather, so that dissolution and decomposition are not caused but it moreover becomes an elevated temperature also at the temperature exceeding 3000 degrees C etc. demonstrates the property which was excellent as high temperature materials, in order to improve the property of this graphite further, sinking copper or other metals into a graphite is performed. Sinking in to a copper graphite material is especially widely carried out industrially for the purpose of electrical conductivity, thermally conductive improvement, improvement in \*\*\*\*\* kinesis, an improvement of a mechanical property, reduction of gas transparency, etc.

[0003] However, generally, when wetting was bad and sank in into pore to a graphite, it did not stick to a cell wall enough, but sinking-in unevenness arose microscopically, and the copper melt had the trouble that the result above-mentioned property improvement could not fully attain.

[0004] As one policy which abolishes sinking-in unevenness, JP,51-24525,A is taught using the copper alloy which added Ti, Zr, or Si as a wetting agent for sinking in to baked-carbon material.

[0005] Moreover, although relevance is thinner than this, manufacture of the metal sinking-in carbon slider material which sinks in antimony and antimony copper after sinking in, pyrolyzing this metal salt beforehand using the salt of the metal of the kind made into the wettability improvement agent of a sinking-in metal and making a metal adhere to a cell wall is taught to JP,61-136644,A.

[0006]

[Problem(s) to be Solved by the Invention] However, in order that the copper sinking-in carbon material concretely indicated by above-mentioned JP,51-24525,A may give \*\*\*\*\* kinesis to the so-called low baking block material of the bulk specific gravity calcinated at about 1200-1500 degrees C, comparatively a lot of copper (about 1083 degrees C of melting points) will be sunk in, and the feature as lightweight high temperature materials of a carbon material will be spoiled. And although Sn, Sb, etc. are added in order to reduce sinking-in temperature, and to raise \*\*\*\*\* kinesis, it may be subtracted rather at improvement of properties other than these end \*\*\*\* kinesis. Moreover, having un-arranged [ of making difficult separation with the sinking-in metal material after sinking in, and carbon material ], since compatibility of Ti with carbon was too high relatively.

[0007] For this invention persons to solve the trouble of the above-mentioned conventional technology and be precise and lightweight, and obtain the copper sinking-in graphite material which was moreover excellent in handling nature The result of wholeheartedly examination, When the isotropic graphite which demonstrates the property which had the homogeneous macrostructure, was precise, divided as

high temperature materials, and was excellent is used, and each compatibility with copper and a graphite carries out selection use of the high element group of a certain kind as a copper additive Could sink in copper uniformly and maintaining the feature as lightweight high temperature materials, it finds out that the copper sinking-in graphite material excellent also in handling nature is obtained, and came to complete this invention.

[0008]

[Means for Solving the Problem] Bulk density is three or more 1.76 Mg/m, and this invention namely, more than 70 volume % of the open pore of isotropic graphite material with the rate of an open pore and average pore radius precise on the organization target which is 5 or 18 volume % and 0.1, or 2.5 micrometers, respectively which were measured by method of mercury penetration Sinking-in processing is carried out by the copper alloy to which at least one sort of elements 1 or 7 % of the weight, and the remainder which are chosen from the element group whose standard enthalpy of formation ( $\Delta H$  degree) by the reaction with a graphite and copper is -50 or less kJs per mol, respectively change from copper substantially. And it is the copper sinking-in graphite material characterized by making bulk density after sinking in into three or less 3.0 Mg/m.

[0009]

[Function of the Invention] In order that this invention persons might study the mechanism of sinking [ of the copper to the inside of graphite pore ] in, they formed the hypothesis by theoretical consideration first, and inquired by the scientific technique of subsequently verifying this hypothesis by experiment. That is, the integrity of sinking [ of the dissimilar material to a certain matter ] in checked the reactivity of copper and a graphite, and various elements, and compatibility first based on view of being deeply related to sinking-in material, the reactivity between sunk in material, and compatibility. The standard enthalpy of formation of the reaction between these elements was shown in drawing 1 . The source of the data in drawing is North-Holland Co. \*\* and Cohesion. in It is the numeric data indicated by Metals.

[0010] Subsequently, out of each element in drawing, each standard enthalpy of formation of a reaction with a graphite and copper was low, namely, the candidate of an assistant who has the integrity of copper sinking in raised formed the hypothesis that it was a negative-number value and an absolute value was also a high element, and tried verification by experiment.

[0011] Consequently, it has checked having relation so that the enthalpy value of drawing 1 and the mechanism of sinking in may explain below.

[0012] That is, although its reactivity with copper is low although the reactivity with a graphite is high, and Ti is advantageous in respect of the interface adhesive strength of generation and CuTi of Ti carbide, and a graphite for this reason, since generation of the carbide in the opposite side surface section becomes superior, sinking in to the in-house back of CuTi is checked, or un-arranging -- separation with the sinking-in metal material after sinking in and graphite material becomes difficult -- arises, for example. On the other hand, if each standard enthalpy of formation with a graphite and copper is low, osmosis to the in-house back of carbide generation, interface adhesive strength, and sinking-in material will be good, and, moreover, will become easy [ separation with the sinking-in metal material after sinking in and graphite material ]. And the threshold of the enthalpy of a reaction with the graphite and copper with which these operations are obtained is [ about ]. -It turns out that it is 50 kJ/a mol. The elements suitable for this condition are Sc, Y, Zr, La, and Hf.

[0013] The bulk density used by this invention by carrying out selection use of the sinking-in assistant of such conditions is three or more 1.76 Mg/m. And are very precise on an organization target [ as / whose rate of an open pore and average pore radius which were measured by method of mercury penetration are 5 or 18 volume % and 0.1, or 2.5 micrometers, respectively ]. Therefore, sinking in can sink in copper to a large quantity and the in-house back also to very difficult isotropic graphite material, copper sinking-in graphite material of a lightweight and precise organization called three or less 3 Mg/m in bulk density also considering rate of impregnation as more than 70 volume % is obtained, and, moreover, the handling nature after sinking in also becomes good. In addition, when the above operations are not fully obtained but exceeded 7 % of the weight, the amount of a sinking-in assistant

was specified as 1 - 7 % of the weight at less than 1 % of the weight, because separation with the sinking-in copper alloy after sinking in and copper sinking-in graphite material became difficult.  
[0014]

[Detailed explanation of the composition of invention] After the isotropic graphite material used by this invention usually adds and kneads binders, such as a pitch, to the aggregates, such as corks \*\*\*\* pressing, such as between the colds, is performed and the need is accepted at baking and graphitization. Sink [ pitch ] in, It consists of the material which makes a principal component the material or \*\*\*\*\* which passed through processes, such as re-baking, resin sinking in, and high-grade-izing, and which consists only of carbon substantially, and isotropic graphite material, such as the so-called graphitization article which includes sinking-in articles, such as a pitch sinking-in article and a resin sinking-in article, is included. Especially as these isotropic graphite material, it is desirable that a different direction ratio uses 1.2 or less isotropic high graphite material. Here, it means that the average of the ratio of the peculiar electric resistance planned in the direction [ in / graphite material / that a different direction ratio is 1.2 or less ] which makes a right angle arbitrarily is 1.2 or less.

[0015] The average pore radius of the isotropic graphite material as used in the field of this invention can be determined as a radius value (micrometer) equivalent to one half of the accumulation pore capacity (m<sup>3</sup>/Mg) measured by method of mercury penetration, and the rate of an open pore can be calculated by  $x(\text{bulk density}) (\text{total pore capacity}) \times 100$ . Here, total pore capacity (m<sup>3</sup>/Mg) says accumulation pore capacity when a pressure reaches to the maximum pressure defined beforehand, for example, 98MPa(s). Moreover, it means that the numeric value shown that rate of impregnation is more than 70 volume % by I (%) of formula:  $I = 100 \text{ G/PD}$  is 70 or more. However, the weight [ actual measurement / of the volume of the open pore of isotropic graphite material / (m<sup>3</sup>) ] (Mg) of the copper alloy into which P sank into and the true density (Mg/m<sup>3</sup>) of copper alloy sinking-in material and G actually sank in D is expressed. That is, I value shows the volume rate of the sinking-in copper alloy occupied to an open pore.

[0016] For example, using a proof-pressure container, sinking in puts a copper alloy into ceramic containers (crucible), such as a product made from carbon, and puts isotropic graphite material into ceramic containers (SAGA), such as a product made from carbon, inserts these crucibles and SAGA in a proof-pressure container, subsequently to temperature higher than the melting point of a sinking-in copper alloy raises the inside of a container, and carries out pressurization sinking in. Number MPa or about 150 MPas, and sinking-in time of welding pressure are preferably good about 1 to 60 minutes at 30 or about 60 minutes.

[0017] In addition, it means that the unescapable impurity may be contained in the copper which constitutes the remainder, saying "the remainder consists of copper substantially" about composition of the copper alloy used in this invention.

[0018]

[Example]

The nature [ of precise ] isotropic graphite material made from example 1 Oriental Carbon (bulk density 1.82 Mg/m<sup>3</sup>, the rate of an open pore of 13.7%, average pore radius of 1.5 micrometers) was held in the proof-pressure container, it carried out pressurization sinking in of the copper which added 7% of the weight of Zr fused at 1150 degrees C by the pressure of 12MPa(s) by N<sub>2</sub> gas for 1 hour, and copper sinking-in graphite material was obtained.

[0019] The bulk density of rate of impregnation and the obtained copper alloy sinking-in material, the gas permeability of copper alloy sinking-in material, and the handling nature after sinking in were summarized in Table 1.

[0020]

[Table 1]

実施例 比較例 番号	含浸助剤	含浸率 (%)	かさ密度 (Mg/m <sup>3</sup> )	ガス透過率 ( $\times 10^{-4}$ · cm <sup>2</sup> /s)	含浸後の取扱い (含浸材と被含 浸材との接着)
実施例 1	Zr 7%	89	2.84	4	接着は多少ある が取り出しは容 易である
対照例 1	なし	67	2.75	200	接着は全くない
比較例 1	Ti 1%	89	2.84	2	接着力が非常に 強く取り出しが 困難である

[0021] Copper sinking-in graphite material was obtained like the example 1 except not adding Zr in example of contrast 1 copper.

[0022] Copper sinking-in graphite material was obtained like the example 1 except having added 1% of the weight of Ti in example of comparison 1 copper.

[0023] Moreover, the thermal conductivity in the room temperature of the isotropic graphite base material used in this invention article concerning an example 1, the example 1 of contrast, the example 1 of comparison, and the example 1, 200 degrees C, 400 degrees C, and 800 degrees C was measured with the laser flash method. The result is shown in Table 2.

[0024]

[Table 2]

処理 温度 (°C)	実施例 1	比較例 1	対照例 1	実施例 1 で 使用した 黒鉛基材
	銅-ジルコニウム	銅-チタン	銅	
室温	190	150	120	140
200	150	130	130	120
400	130	120	110	100
800	110	110	100	70

1) 熱伝導率はレーザーフラッシュ法で測定した。

2) 単位は、W/(m·K)である。

[0025] It not only raises an adhesive property with graphite material, but by alloying Cu with Zr, it can obtain the graphite material of high thermal conductivity.

[0026]

[Effect of the Invention] According to this invention, it has a homogeneous macrostructure and is precise, and it is precise, being able to sink in copper uniformly and maintaining the feature as

lightweight high temperature materials, even if it uses the isotropic graphite which demonstrates the property which divided as high temperature materials and was excellent, and the copper sinking-in graphite material excellent also in handling nature is obtained.

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**CLAIMS**

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[Claim(s)]

[Claim 1] Bulk density is three or more 1.76 Mg/m, and more than 70 volume % of the open pore of isotropic graphite material with the rate of an open pore and average pore radius precise on the organization target which is 5 or 18 volume % and 0.1, or 2.5 micrometers, respectively which were measured by method of mercury penetration Sinking-in processing is carried out by the copper alloy to which at least one sort of elements 1 or 7 % of the weight, and the remainder which are chosen from the element group whose standard enthalpy of formation by the reaction with a graphite and copper is -50 or less kJs per mol, respectively change from copper substantially. And copper sinking-in graphite material characterized by making bulk density after sinking in into three or less 3.0 Mg/m.

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